Magnetic instability induced by tunnel current in single Co nanoparticles

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Measurements of magnetic hysteresis loops in single Co nanoparticles at dilution refrigerator temperatures are presented. The nanoparticles are in electric contact with bulk Al leads via tunnel junctions. The tunnel current versus magnetic field displays a magnetic hysteresis loop. The magnetic switching field is reduced by current, and the magnetization of the nanoparticle can be switched by applying a voltage pulse, demonstrating that the magnetic stability of the nanoparticle is diminished by electron transport. Additional transitions and random switches of magnetization can be driven by large currents.

The field of spintronics has shifted from the study of fundamental physics to technological devices in past decades and holds the promise for future application such as magnetic random access memory. Driven by trends towards high density storage and miniaturization of devices, the size of the magnetic unit cell keeps shrinking. Recently, studies on spintronics have been driven to the molecular-scale level[1–4], which offers the possibility of exploiting quantum effects. Most spintronic applications rely on the magnetic order of the nanomagnets being stable with time. However, since the anisotropy energy holding the magnetic moment of nanomagnets decreases with size, random switches of magnetization may be triggered by perturbations. It is well known that thermal fluctuations can induce the random flipping of the magnetization in nanomagnets, the effect normally referred to as superparamagnetism. To overcome the blocking temperature, extensive studies have been performed and some proposals have been raised such as using exchange bias[5,6] to beat the superparamagnetism. Similar to thermal fluctuation, current incident from normal metal carrying random electron spins can also induce instability in ferromagnets[7–12].

In this work, we study effect of tunnel current on the magnetization of single Co nanoparticles at mK temperature range. We first reproduce some results from Refs. 13 and 14. Then, we find that tunnel current from normal leads can greatly reduce the magnetic switching field of Co nanoparticles. We confirm that this reduction in switching field is not due to the Joule heating. Thus, the electrons tunneling from normal metal can randomize the magnetization of Co nanoparticles. Our results point out a challenge in spintronics open for further explorations.

Our samples consist of Co nanoparticles tunnel-coupled with two Al leads via alumina barriers. Figs. 1A and 1B, show the micrograph of a typical device and the fabrication process,[15,16] respectively. A polymethilmetachryllate (PMMA) bridge over a SiO_2 substrate, is defined by electron beam lithography. First, we deposit 10nm of Al and 1.5-3nm of Al_2O_3, along direction 1. Then we oxidize the sample at room temperature in O_2 at 3mPa, for 30s. Next, we deposit 0.5nm of Co, 1nm-1.5nm of Al_2O_3, followed by 10nm of Al, along direction 2. At such a small thickness (0.5nm), isolated Co nanoparticles are formed 1-4nm in diameter and spaced

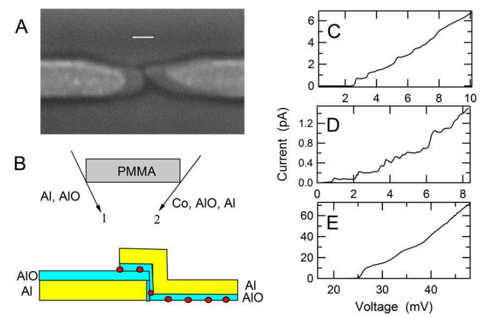

FIG. 1. A: Scanning electron micrograph of a typical sample. The scale bar indicates 0.2μm. B: Sketch representing the sample fabrication process. C-E: Current versus voltage in samples 1-3 (top to bottom), at 60mK and 0T.

by 2-5nm.[13] The current-voltage characteristics, $I(V)$, and the tunnel spectrum, $\mathrm{d}I(V)/\mathrm{d}V$, at $T = 15$mK and zero applied magnetic field, are shown in Fig. 1C-E. Both the Coulomb blockade region and the discrete energy levels are well resolved.

Fig. 2A displays the magnetic field dependence of the energy levels, for decreasing and increasing magnetic fields, showing symmetric hysteresis about zero field. The base temperature varies from 15mK to 60mK in this work depending on the magnetic sweep rate. All of the levels are discontinuous at the switching field $|B_{sw}| \approx 0.3$T, indicating that they belong to the same nanoparticle. Note that the magnetic field dependence of the energy levels varies from level to level. This variation is known to be related to mesoscopic fluctuations (a few percent) of the magnetic anisotropy energy.[13,14,17–19] The inset in Fig. 2B shows the energy levels versus magnetic field in strong field. As in previous work,[13,14] most of the levels shift in parallel. The slope corresponds to the g-factor of 1.0. The energy of these levels decreases with magnetic field, indicating that a minority electrons initially tunnels off the nanoparticle.[14] There was a small Q_0 shift between Figs. 2A and 2B.

The switching field at zero tunnel current is measured in the Coulomb blockade state. We set the voltage to 2.4mV and measure the magnetic hysteresis loop. In the vicinity of the switching field, we follow the path indi-

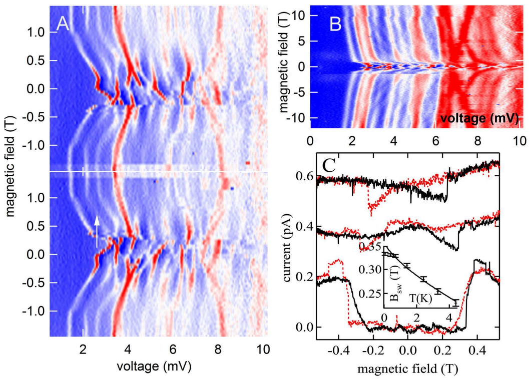

FIG. 2. Sample 1. A and B: Differential conductance versus magnetic field and voltage at base temperature. Top and the bottom panel in A correspond to decreasing and increasing magnetic field, respectively. Blue/Dark (red/bright) regions correspond to low (high) conductance. C: Current versus magnetic field at voltage 2.4mV, showing the hysteresis loop at $T = 60$mK, 2.4K, and 4.4K (bottom to top), with current offsets 0.2pA (for clarity). Black/Full (red/dashed) line correspond to increasing (decreasing) field. Inset: Switching field versus temperature.

cated by the white arrow in Fig. 2A. At low temperatures, if the magnetic field is just below the switching field, the nanoparticle will face Coulomb blockade. As the magnetic field is increased and reaches the switching field, a transition to a current-carrying state takes place. The magnetic hysteresis loop for three different temperatures is displayed in Fig. 2C, with the inset showing the temperature dependence of the switching field averaged over ≈30 field cycles. The error bars indicate the standard deviation of the switching field. The decrease of the switching field with temperature, accompanied by an increase in the standard deviation, indicates thermally activated switching.[20–22] By fitting the temperature dependence of the intrinsic switching field and the standard deviation, to the Néel-Brown model of magnetic reversal,[23,24] following the procedure in Ref. 22, we obtain B_0 =0.359±0.036T for the switching field at zero temperature and E_B =20.6±2meV for the anisotropy energy barrier. The fit of the switching field is shown by the solid line in the inset in Fig. 2C. We can estimate the "thermally activated" spin S as $E_B/(g\mu_B B_0) \approx 1000$, and the corresponding magnetic volume $V = (2.4\text{nm})^3$,[22] which is in agreement with the expected particle size.

Next, we investigate the effects of the tunnel current on magnetic switching at $T = 60$mK. Fig. 3A shows that the magnitude of the switching field decreases with current in sample 1. The phase diagram in Fig. 3B, corresponding to the increasing-field branch, displays the $I(B, V)$ dependence in detail. During the measurement of $I(B, V)$, the magnetic field is cycled fast, while the voltage is varied slowly, from 10mV to zero and then back to 10mV. We subtracted a smooth, field independent current background in order to enhance the magnetoresistance contrast. At the Coulomb blockade threshold, the switching

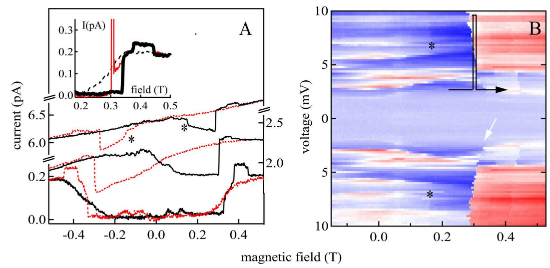

FIG. 3. Sample 1 at base temperature. A: Current hysteresis loops at voltage 2.4mV, 5.2mV, and 10mV (bottom to top). Black/Full (red/dashed) line correspond to increasing (decreasing) field. Inset: Hysteresis loops zoomed-in near the switching field. Full lines correspond to increasing field. Red/Thin (black/thick) correspond to with (without) the voltage pulse. Dashed line corresponds to decreasing field. B: Current versus increasing magnetic field and voltage. The white arrow points to the switching field versus voltage. The dark arrow represents a crossing of phase boundary induced by voltage. The stars indicate current driven magnetic transitions.

field (B_{sw}), indicated by the arrow, decreases with current at a rate $\mathrm{d}\,lnB_{sw}/\mathrm{d}I = 5\%\text{pA}^{-1}$, where I is the tunnel current measured just before the switching. This rate is a measure of the magnetic response of the particle to the tunnel current, and will be further referred to as current susceptibility. The current susceptibility decreases rapidly as a function of I. At a bias voltage of 10 mV, where the tunnel current before the transition is close to 6pA, the switching field is reduced by ≈15% compared to the value at zero current. Meanwhile, the current-susceptibility is reduced to $0.6\%\,\text{pA}^{-1}$, an order of magnitude suppression compared to that near zero tunnel current. The smooth background pattern in Fig. 3B arises from the motion of the levels with the magnetic field. The phase diagram for the decreasing-field branch (after reflection about zero magnetic field) as well as for negative bias voltage, present similar behavior.

We have studied four samples at dilution refrigerator temperatures in detail. One sample does not display any reduction in the switching field with current near the Coulomb Blockade threshold, the other samples display a switching field reduction similar to that above (Fig. 4A inset, B-C). In sample 3, the tunnel resistance at high voltages is four times smaller than in sample 1. At $T = 60$mK, the current-field hysteresis loop versus voltage is shown in Fig. 4B, and the phase diagram is displayed in Fig. 4C. Near the Coulomb blockade voltage threshold, the switching field decreases rapidly with current, with the initial current susceptibility $\mathrm{d}\,ln\,B_{sw}/\mathrm{d}I \approx 4\%\,\text{pA}^{-1}$. Similar to sample 1, the current susceptibility drops rapidly with the tunnel current. In both samples, when the tunnel current is large, the hysteresis loop starts to display additional magnetic transitions, indicated by the stars in Figs. 3 and 4B-C. Those transitions are current-induced, since they are absent near the Coulomb blockade conduction threshold. In sample

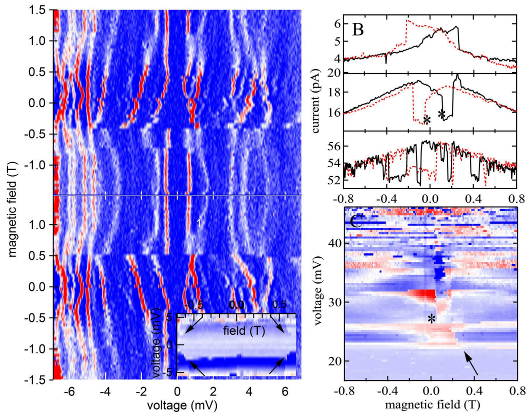

FIG. 4. A: Sample 2 at base temperature. Differential conductance versus magnetic field and voltage. Top and bottom panel correspond to decreasing and increasing magnetic field, respectively. Blue/Dark (red/bright) regions correspond to low (high) conductance. Inset: Current versus decreasing and increasing magnetic field and voltage. The arrows point to the switching field versus voltage. B and C: Sample 3 at base temperature. B: Current hysteresis loops, measured at 26mV, 32mV, and 44mV (top to bottom). Black/Full (red/dashed) line correspond to increasing (decreasing) field. C: Current versus increasing field, as a function of voltage. The black arrow points to the switching field versus voltage. The stars indicate current driven magnetic transitions.

3, the magnetic hysteresis disappears completely at the blocking voltage (∼38mV), above which, the particle is in a superparamagnetic-like state, exhibiting telegraph noise, as seen in Figs. 4B-C. We measured the phase diagram of sample 2 at 4.2K, and found that the the current susceptibility is suppressed compared to that at $T =$ 60mK. Near the Coulomb blockade voltage threshold at $T = 4.2K$, we obtain $\mathrm{d}\, ln\, B_{sw}/\mathrm{d}I \approx 0.2\%\,\mathrm{pA}^{-1}$. Thus, as the temperature and the tunnel current approach zero, the current susceptibility is strongly enhanced.

Now, we present the current induced magnetic switch in sample 1. We vary the parameters following the black line in Fig. 3B. The initial voltage is 2.4mV, below the Coulomb blockade threshold. This time, we cross the phase boundary by applying a 10mV voltage pulse at 0.3T, well before the positive switching field and then compare it with the ordinary hysteresis loop. The inset in Fig. 3A displays the two loops in the vicinity of the switching field. The black/thick full line displays current versus increasing field, in the absence of the pulse. The dashed line displays current versus decreasing field, and the red/thin full line displays current versus increasing field, in the presence of the pulse. The voltage pulse switches the nanoparticle from the Coulomb blockade into the current carrying state. When the voltage is set back to 2.4mV, the current settles around the magnetically reversed state, showing that the state of the nanoparticle has been switched. Magnetic switching by voltage pulse has been tested on three hysteresis loops and at the negative switching field as well, with the voltage-driven switching being confirmed every time.

One explanation that current can reduce the switching field is sample heating. However, it is not the case here. In sample 1, the switching field is reduced by ≈10% at 6.3mV. If the reduction was the result of sample heating, then the sample temperature would have to be ≈2K, from the inset in Fig. 2C, to explain the reduction. The width of the tunnel resonances measures the electron temperature in the leads directly. Many tunnel resonances in samples 1 and 2 remain very sharp at these voltages near zero magnetic field, indicating electron temperature <0.2K. Thus, heating in the leads cannot be responsible for the reduction of the switching field by tunnel current.

The absence of Joule heating suggests an interpretation that sequential electron transport randomizes the Co nanoparticle magnetization. The randomization of magnetization can be attributed to spin-shot noise[11] or magnetization random walk in response to sequential electron transport[12]. The current driven additional switches and telegraph noise state are analogous to superparamagnetic state. Thus, electron tunneling from normal leads can induce instability in the magnetic moment of a Co nanoparticle. In future research, gate voltage dependence of the magnetic hysteresis loop will be measured, to determine if nonequilibrium particle-hole excitations are responsible for the hysteresis loop narrowing.

In conclusion, tunnel current from normal metal leads reduces magnetic switching field of single Co nanoparticles without raising the temperature of the leads. The magnetization of the nanoparticle can be switched by applying a voltage pulse when magnetic field is below the switching field. Additional transitions and random switches of the magnetization in Co nanoparticles can be driven by large current. These effects suggest that tunnel current can destroy the stability of the magnetization in Co nanoparticles, similar to the phenomenon of superparamagnetism in response to thermal fluctuations. The effects presented in this paper introduce a challenge in the miniaturization of spintronic devices open for exploration.

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